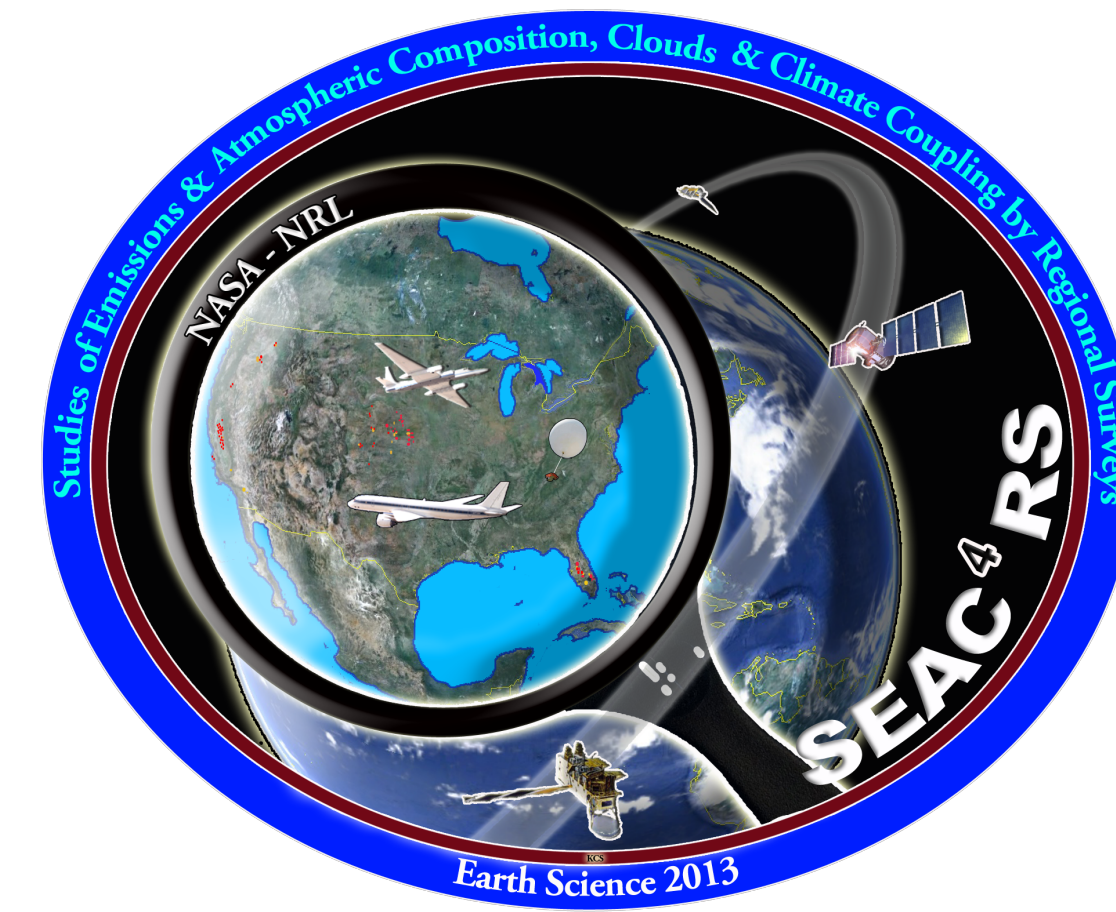




Size-resolved, Subsaturated, Aerosol Hygroscopicity During the SEAC⁴RS Field Campaign

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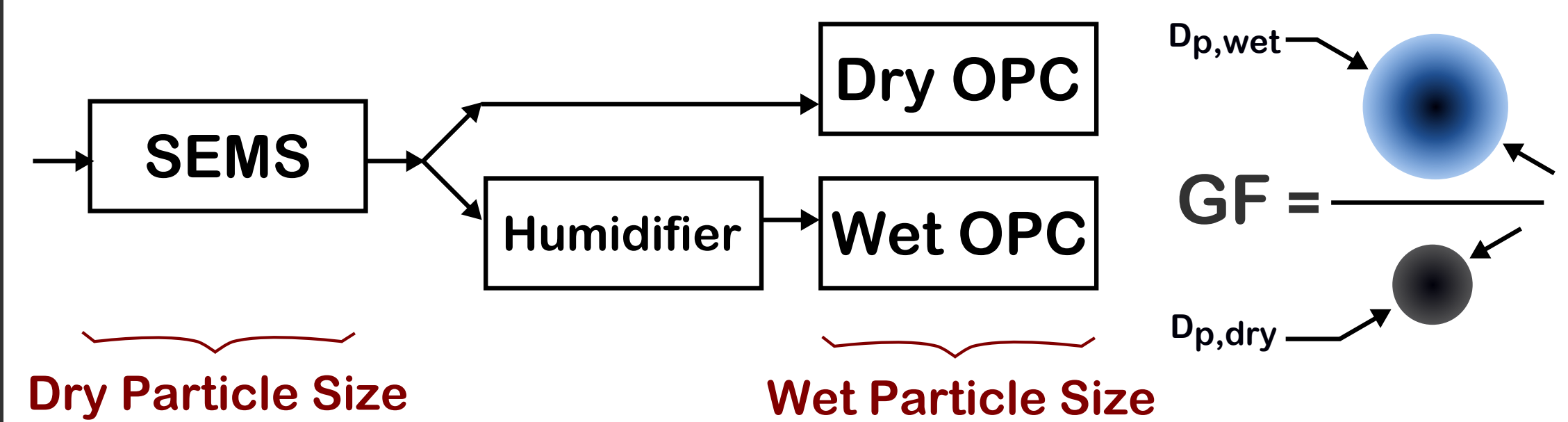


The DASH-SP on the NASA DC-8

Aerosol particles can scatter and absorb solar radiation and serve as cloud condensation nuclei (CCN), thereby influencing cloud properties and precipitation. The radiative and CCN-relevant properties of particles depend on their hygroscopicity. As particles are exposed to changing relative humidity (RH), they undergo a change in their size owing to hygroscopic growth (via water-uptake) with the magnitude of this growth dependent on their chemical composition.

The Differential Aerosol Sizing and Hygroscopicity - Spectrometer Probe (DASH-SP; Brechtel Mfg. Inc.; Sorooshian et al., 2008) is capable of rapidly measuring size-resolved hygroscopic growth factors (GFs) of ambient aerosol particles. GF values are defined as the ratio of a particle's diameter when exposed to elevated RH values (typically ~70-95 %) vs the particle's diameter when dried to <15 % RH. The polydisperse aerosol sampling stream is first size selected using a Scanning Electrical Mobility and Sizer (SEMS) to create a monodisperse aerosol flow at a single diameter between 175 - 350 nm. A flow splitter separates the monodisperse aerosol sample into two paths with equivalent total residence times through the "dry" and "wet" optical particle counters (OPCs). The humidifier has the ability to quickly equilibrate the sample flow to the desired RH before reaching the OPC without diluting the sample with dry/filtered air.

The DASH-SP instrument has much faster time resolution than widely used alternative hygroscopicity measurement techniques, including the HTDMA. Single scans can be acquired in as little as 1 s. The average uncertainty associated with DASH-SP GF measurements is ± 4.3 %. A range of scans covering $D_{p,dry}$ sizes 175 - 350 nm at a single RH can be completed in under 5 minutes with the majority of the time being required by the SEMS between size changes. Performing replicate scans at each dry size before switching greatly increases the number of scans collected in a given time.



Emission Source Impacts on Water Uptake

Figure 1. Growth factors as a function of relative humidity are plotted for flights RF11 (fresh biomass burning, BB) and RF12 (aged BB). The average growth factors for each of the entire flights are shown with lines and whiskers (standard deviations). Marker size is representative of $D_{p,dry}$, ranging between 200-275 nm. Average growth factors are greatly suppressed in the fresh emissions and increase with age at all relative humidities. Further investigation will be conducted to determine the response of growth as a function of air mass age at fixed RH and $D_{p,dry}$ values.

Table 1. Average growth factors, at various measurement RH values, and dry refractive index (Dry RI; real component only), as a function of size (200-275 nm) for aerosols sampled during all available flights. Standard deviations are shown in parenthesis. Measurements are grouped into emission source categories. The BB rich emission sources have significantly decreased average growth factors at all size/RH combinations, while biogenic volatile organic carbon (BVOC) rich emission sources had only moderate decreases in average GF - at lower RH. The refractive index of the dried aerosols only varied slightly between the emission sources and negligibly between different sizes of a single emission source.

Subsaturated Aerosol Growth Factors as a Function of Size and RH						
Air mass	$D_{p,dry}$ (nm)	Sample Relative Humidity				Dry RI
		75%	80%	85%	90%	
Non-smoke, Non-biogenic	200	1.33 (0.17)	1.45 (0.15)	1.50 (0.13)	1.52 (0.19)	1.541 (0.013)
	225	1.27 (0.13)	1.38 (0.13)	1.42 (0.16)	1.53 (0.14)	1.542 (0.010)
	250	1.29 (0.13)	1.36 (0.13)	1.40 (0.14)	1.50 (0.14)	1.546 (0.012)
	275	1.35 (0.09)	1.38 (0.16)	1.45 (0.14)	1.38 (0.13)	1.544 (0.011)
	275	1.35 (0.09)	1.38 (0.16)	1.45 (0.14)	1.38 (0.13)	1.544 (0.011)
Biomass Burning	200	N/A	1.12 (0.16)	1.28 (0.15)	1.13 (0.15)	1.536 (0.011)
	225	1.11 (0.12)	1.16 (0.15)	1.18 (0.17)	1.40 (0.11)	1.552 (0.012)
	250	1.20 (0.10)	1.14 (0.10)	1.19 (0.12)	1.26 (0.13)	1.556 (0.015)
	275	1.25 (0.10)	1.28 (0.08)	1.32 (0.11)	1.32 (0.08)	1.551 (0.007)
	275	1.25 (0.10)	1.28 (0.08)	1.32 (0.11)	1.32 (0.08)	1.551 (0.007)
Biogenic Emissions	200	1.18 (0.11)	1.29 (0.13)	1.42 (0.10)	1.52 (0.10)	1.537 (0.005)
	225	1.27 (0.09)	1.33 (0.11)	1.41 (0.08)	1.54 (0.09)	1.536 (0.009)
	250	1.28 (0.06)	1.34 (0.09)	1.41 (0.08)	1.53 (0.09)	1.535 (0.007)
	275	N/A	1.19 (0.02)	N/A	1.21 (0.04)	1.543 (0.003)
	275	N/A	1.19 (0.02)	N/A	1.21 (0.04)	1.543 (0.003)

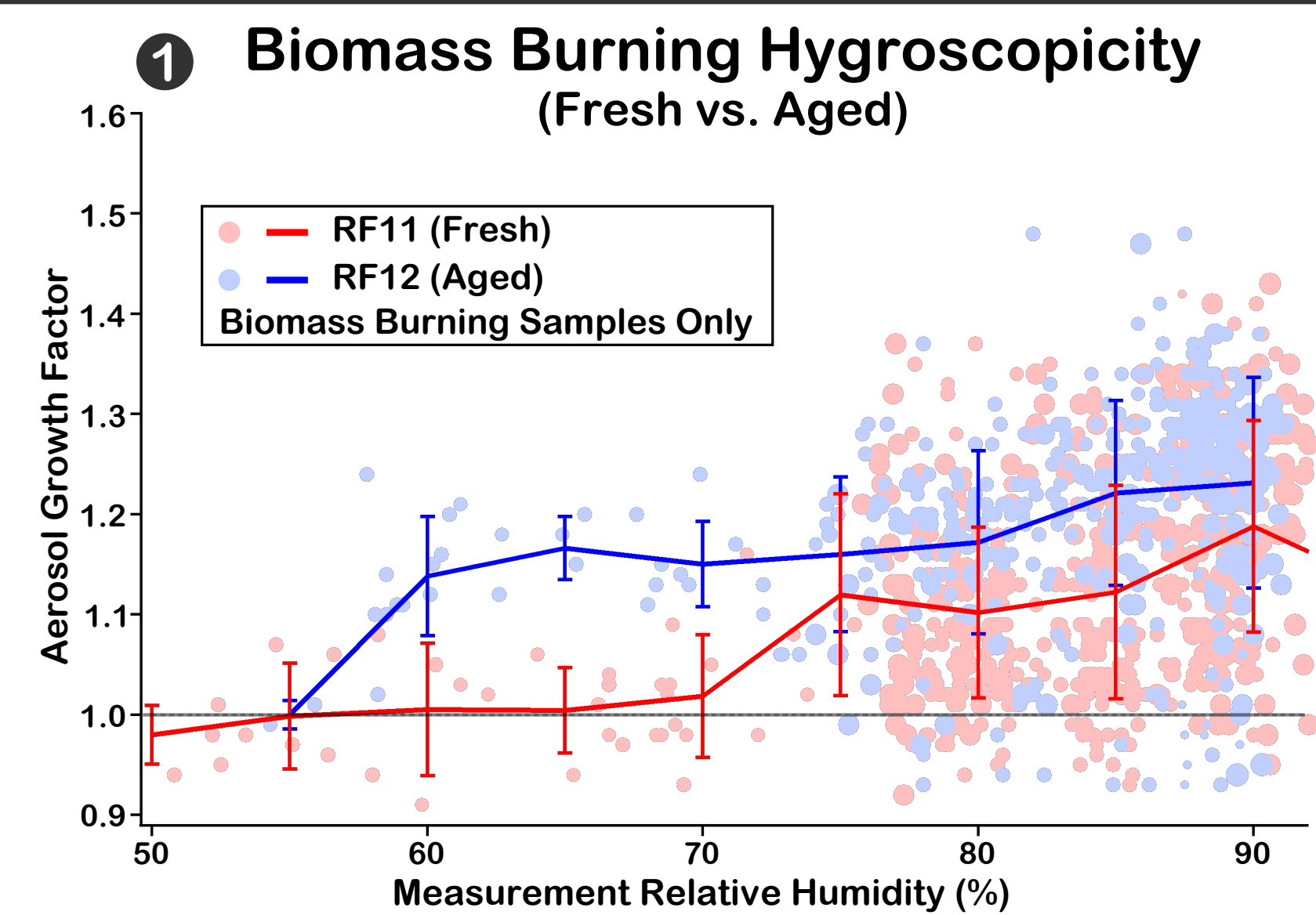
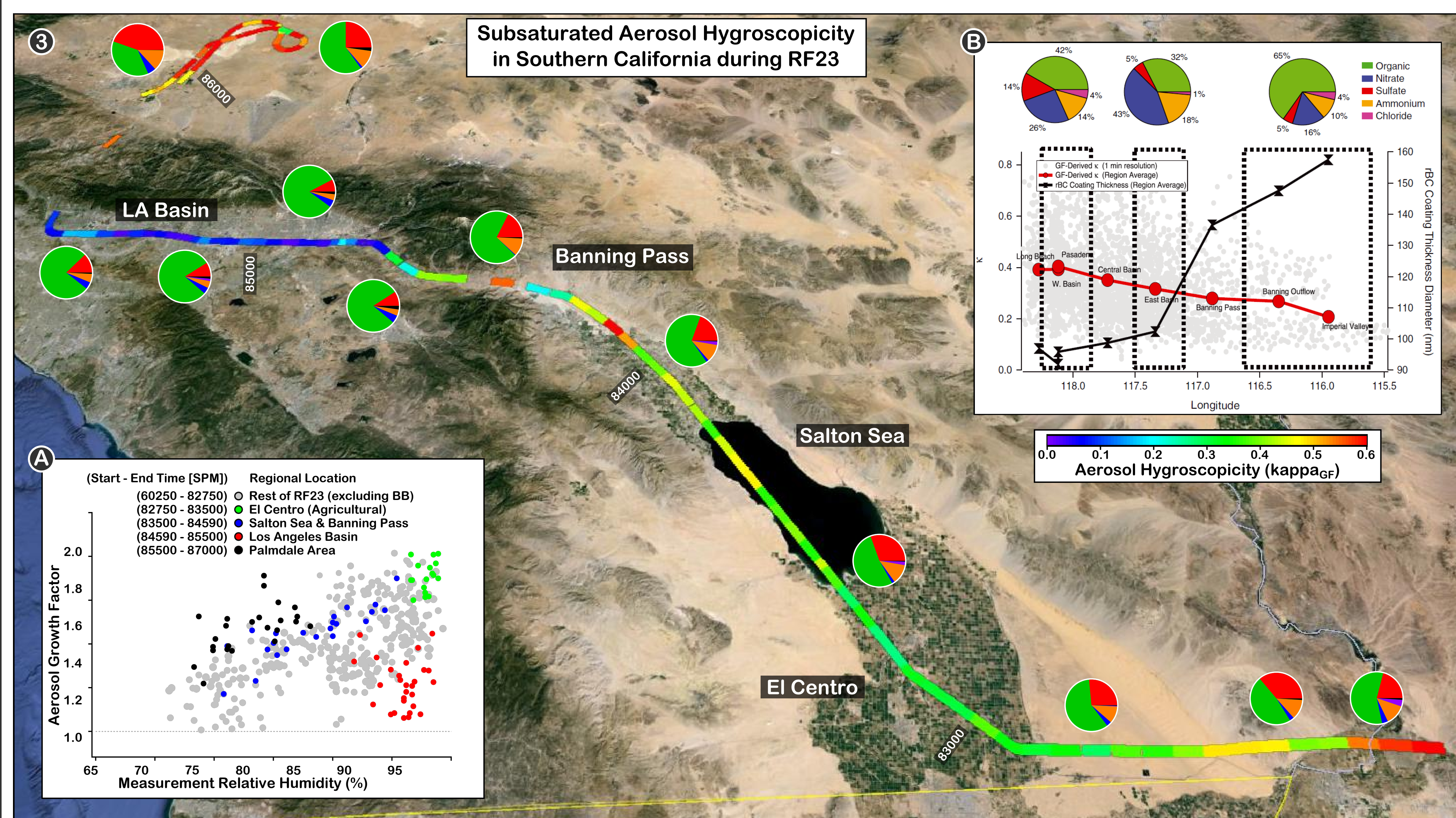
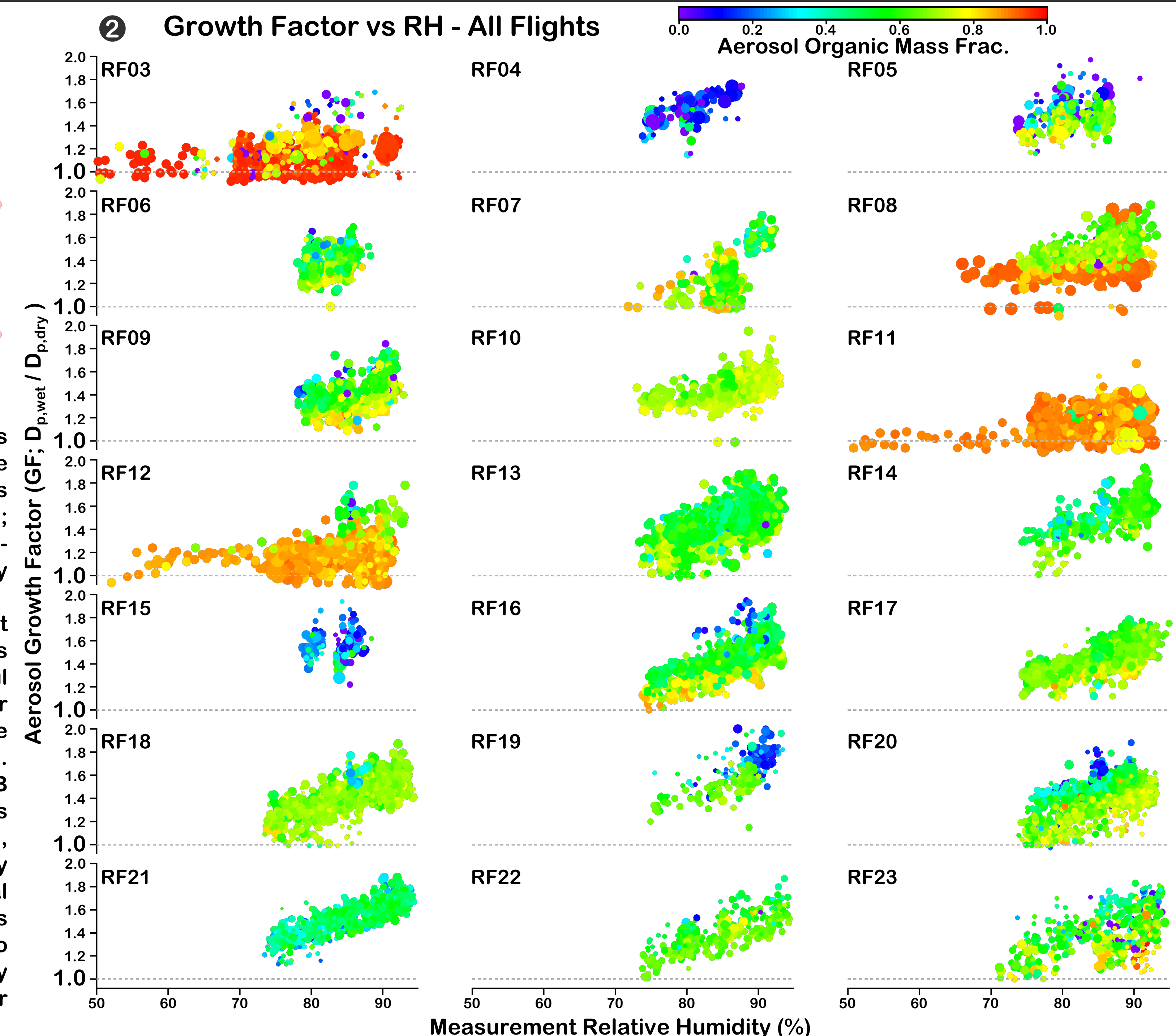


Figure 2 (right). Individual growth factor vs. RH plots are shown for research flights (RF) 3 through 23. The markers are color coded with organic mass fractions (measured by the aerosol mass spectrometer, AMS; prelim. data). Data shown is filtered to remove any in-cloud sampling times and is limited to sample availability coincident between the DASH-SP and AMS.

Hygroscopic growth of aerosols is highly dependent on aerosol chemistry. The percentage of mass composition which is organic material is a useful indicator in aiding to distinguish separate air masses or changes in an air mass over time. Organic aerosols are generally less hygroscopic than inorganic aerosols (e.g. $(NH_4)_2SO_4$ and NH_4NO_3). The organic mass fraction of BB aerosols is very high and flights targeting these sources are easily identifiable in the plot (orange-red; RF-03, -08, -11, -12). In contrast, sampling of urban areas frequently contains a higher concentration of inorganic material and tends to yield more hygroscopic growth (blue colors in plot; RF-04, -15, -16, -19, -20). In between these two extremes, BVOC rich sources tend to have moderately high organic content and produce moderately lower hygroscopic growth factors.



Case Study: Boundary Layer Transect Through the LA Basin

A long, low-level (< 500 mAGL), transect from Yuma, Az., through Banning Pass and the LA Basin, over the San Gabriel Mountains, and finishing in Palmdale, Ca., was conducted at the end of RF23.

In the area near Yuma, Az., the hygroscopic growth was relatively high compared to background samples (kappa between 0.5 and 0.6) and organic mass fractions of ~50%. Agricultural use lands in the El Centro, Ca., area and up through Banning Pass had moderate growth, with kappa values between 0.25 and 0.50, similar to background samples, with total aerosol mass concentrations of less than $4.0 \mu g m^{-3}$.

Entering into the LA Basin, the wind direction changed from out of the East to out of the West and the total aerosol mass concentration spiked to $10.6 \mu g m^{-3}$. The organic mass fraction of aerosols increased to ~84% and kappa decreased to below 0.1. The total mass steadily increased through the basin up to $19.3 \mu g m^{-3}$ before going over the San Gabriel Mountains and down into Palmdale, Ca., where the total mass concentrations returned to below $4.0 \mu g m^{-3}$ and the organic mass fraction decreased to below 45%. Much higher hygroscopic growth was seen in the Palmdale, Ca., area with kappa values between 0.5 and 0.6.

Similar measurements were collected in 2010 during CalNex by Hersey, et al. (2013); however, they report seeing the opposite trends in organic mass fractions and hygroscopic growth. Traveling west to east from the LA Basin, through Banning Pass, and out into the Imperial Valley, organic fractions increased, and hygroscopicity decreased steadily. While their measurements are averaged over 17 flights compared to the single transect performed here, the complete switch in results is of interest and will be investigated further using size-resolved composition data to aid in analyzing the hygroscopic response observed during RF23.

Figure 3. The flight path for the RF23 transect through the LA Basin is plotted over Google Earth and color coded with the hygroscopicity parameter "kappa" (Petters and Kreidenweis, 2007), where a value of zero indicates non-hygroscopic material and increases with increasing hygroscopicity. Aerosol mass composition from the AMS is depicted with pie charts. See inset (b) for composition details. Numbers adjacent to the flight path indicate the time (UTC) in seconds past midnight at that point of the flight. Inset (a) includes the aerosol GF vs. RH response for RF23, color coded by the region of the transect. Inset (b) is a copy of Figure 7 from Hersey, et al. (2013), where similar measurements were made during the CalNex field campaign.

Key Research Interests

- 1) Composite and investigate subsaturated aerosol average growth factors in specific emission source air masses for all flights during SEAC4RS.
- 2) Explore effects of cloud processing on aerosol hygroscopicity of advected air masses as well as impacts on water uptake properties from increases/decreases in photo-oxidation rates in the presence of cloud layers.
- 3) Investigate the effects of aging on biomass burning hygroscopicity and validate sub-1.0 growth factors observed in air masses containing freshly-emitted biomass burning aerosols.
- 4) Analyze agreement between the sub- and super-saturated derived kappa values with an emphasis on biomass burning emissions. Examine compatibility between size-resolved and bulk, subsaturated hygroscopic measurements.
- 5) Determine applicability of using simple mixing rules in subsaturated aerosol growth estimates using size-resolved aerosol chemistry data.

References

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